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Studies of nuclear disintegrations with a coincidence spectrometer

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With 9 figures in the text

Summary

A spectrometer for electron-electron and electron-gamma coincidence measurements is described. Attention is called to some of its applications, namely (i) the study of complex nuclear disintegrations, (ii) determinations of internal conversion coefficients and branching ratios and (iii) measurements of the lifetimes of specified nuclear levels. The experimental difficulties and the limitations encountered are discussed in some detail. The instrument has been used for the study of the following isotopes: Pb^{200}, Pb^{205} and Tl^{200}. Finally, a new method is suggested for the measurement of very shortlived nuclear levels ($\leq 10^{-10}$ sec.) with the aid of the electron-electron coincidence spectrometer and a high-frequency transmitter.

Introduction

Coincidence methods can be applied in many different ways to the study of nuclear disintegrations. In earlier experiments [1] of this kind there was generally either no energy selection at all of the radiation under investigation or only a rough discrimination was provided by means of suitable absorbers. In 1937 Bothe and Maier-Leibnitz [2] developed a single channel electron-gamma coincidence instrument. This consisted of a semicircular spectrometer provided with an additional gamma sensitive detector, situated near the source. A double channel electron-electron coincidence spectrometer was first suggested by Feather [3] in 1940. The instrument is built up of two semicircular spectrometers, operated in parallel, with a common source. Various modifications of the latter instrument, utilizing the semicircular [4] or short lens [5] focusing principles have been developed.[1]

The coincidence counting rate is of course proportional to the number of disintegrations per second. It is easy to see that the maximum source strength is limited by the resolving time of the coincidence circuit. G.M. counters are characterized by a comparatively slow response to the incident electron. Therefore only weak sources can be used and the coincidence counting rate will be inconveniently low. The situation is considerably improved if the G.M. counters are replaced by fast organic scintillation detectors. But, even then, the method is of limited value, since the coincidence counting rate is also proportional to the product of the transmissions in the two channels. The inherent low transmission of the semicircular and short lens spectrometers introduces severe limitations in this respect.

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[1] For a review of various types of coincidence spectrometers see T. R. Gerholm, Beta-Ray Spectroscopes, *Encyclopedia of Physics* (editor S. Flügge), Springer Verlag 1956, Vol. 33, p. 668.

The long lens spectrometer, however, gives, at the corresponding resolution, about ten times higher transmission. The long lens focusing principle should, therefore, be very suitable for an electron-electron coincidence spectrometer. In fact, for an instrument of this kind the coincidence counting efficiency will be comparable to the effective transmission of semicircular or short lens spectrometers. The coincidence counting efficiency is the probability that a coincidence pair emitted from the source will be recorded as a coincidence when the two channels are adjusted to focus the members of this pair.

A coincidence spectrometer making use of the long lens focusing principle in both channels has been built and described by Gerholm [6]. It is the purpose of this paper to discuss the coincidence spectrometer method in more detail and to give an account of a number of different applications of current interest. For some types of measurements the applicability of the method has evidently not been fully realized or at least not been completely explored so far. The limitations and experimental difficulties will also be discussed.

The coincidence spectrometer

Since details of the construction have been published elsewhere [6] only a brief account of the main features of the spectrometer will be given here. As shown in Fig. 1 the instrument consists of two long lens spectrometers, placed end to end. By means of the iron shields in the midplane of the instrument the two halves are made magnetically independent of each other. It is therefore possible to focus, simultaneously, electrons of a certain energy in one half and of a different energy in the other. Thin anthracene crystals cemented onto lucite light guides are used as detectors. The light guides have been given a certain profile in order to increase the light collection efficiency. The signals from the photomultipliers are fed, via preamplifiers to a coincidence equipment of the fast-slow type [7], supplied with a variable delay line. The delay is used to determine the background of random coincidences. This background is measured with a sufficiently large time delay introduced in one of the two channels. The time delay can also be used to determine the lifetimes of specified nuclear energy levels (see below). The bias level of the discriminators in the side channels are adjusted to reject pulses with amplitudes lower than those produced by the electrons focused in the corresponding spectrometer.

The transmission–resolution settings can be varied at will by screwing the B_2-baffles towards or away from the conical surfaces, which are machined stepwise to reduce scattering. The entrance baffles B_1 are kept fixed and only serve to reduce the background of scattered electrons. The baffle system is readily accessible because the flanges, with pole pieces and detectors, swing out on hinges. The spectrometer pressure is generally pumped down from atmospheric to vacuum within a few minutes.

The magnetic field used to focus the electrons in each of the two halves is of "tringular" shape. It can be shown [8] that this particular field distribution gives a high luminosity. This is fortunate, since it provides a compensation for the small dimensions of the instrument. The distance between the source and each detector is only 25 cm, but the spectrometer is nevertheless capable of dealing with rather

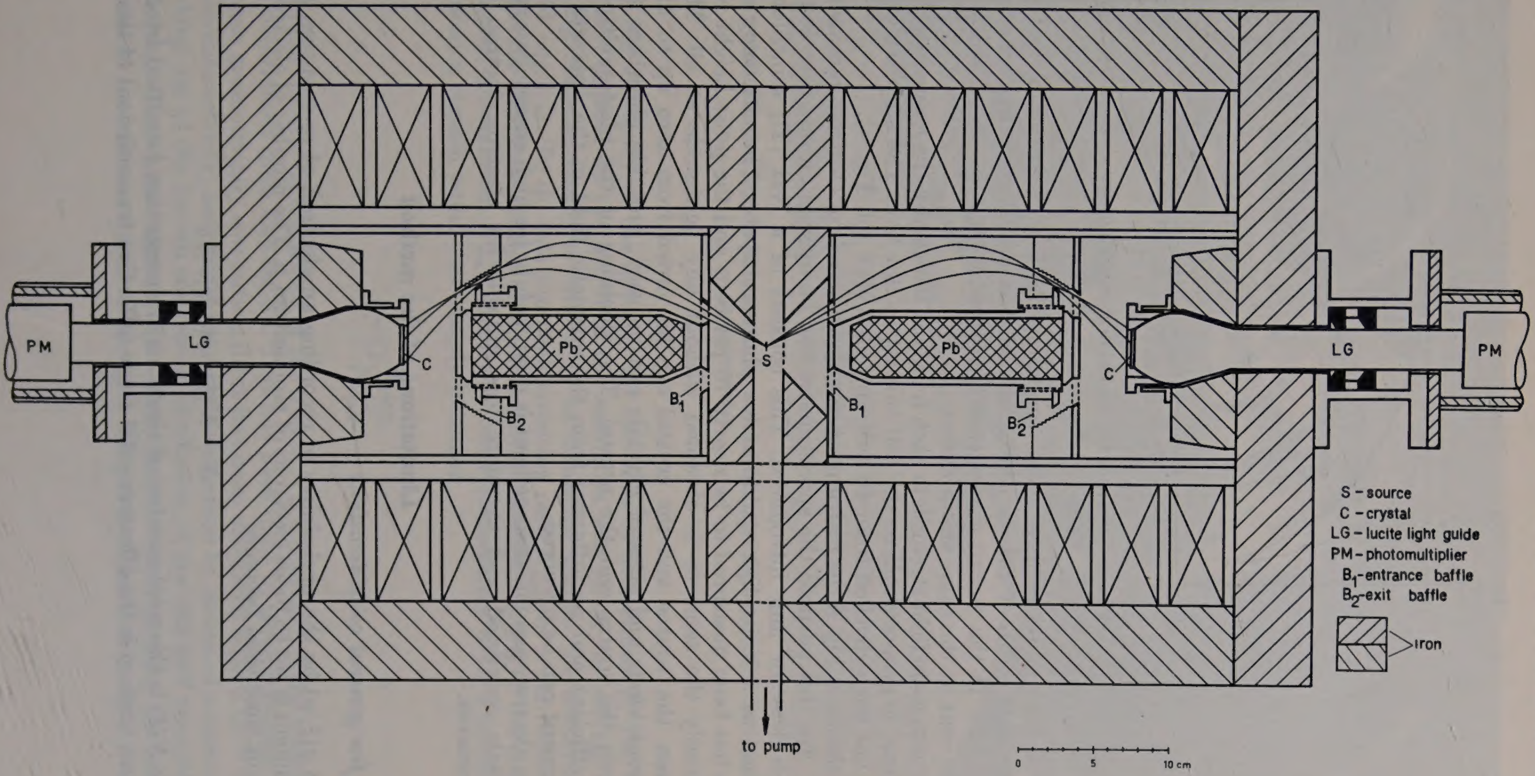

Fig. 1. Electron-electron coincidence spectrometer.

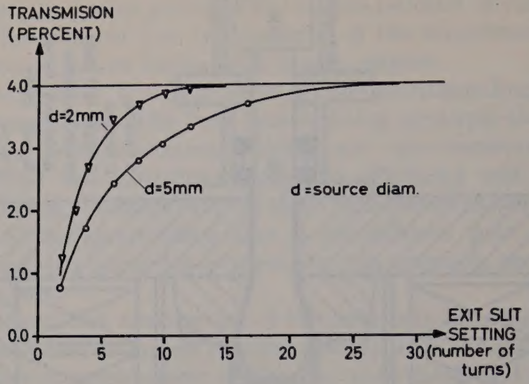

Fig. 2. Effective transmission versus exit slit setting.

large sources maintaining a good resolution. With a source diameter of 0.2 cm a relative half width of 1.3 per cent is obtained at an effective transmission of about 3 per cent. If the source diameter is increased to 0.5 cm the resolution becomes 3.1 per cent at the same transmission.

The magnetic field strength in each half, and hence also the momenta of the focused electrons, is proportional to the coil current in this half. The linearity of the instrument has been checked from 40 keV up to 3.6 MeV (cf. Fig. 4).

As shown in Fig. 6 the instrument can also be used for electron-gamma coincidence work. For this purpose the spectrometer can be opened on one side and a separate gamma-detector unit introduced. This consists of a NaI (Tl) scintillation crystal attached to a lucite light guide and a photomultiplier. The efficiency of the light guide has been measured to be nearly 70 per cent and the pulse height resolution is practically the same as is obtained without any lightguide at all. The distance between the source and the crystal can be varied from zero up to about 5 cm. Different kinds and sizes of crystals can be used. Suitable absorbers are inserted between the source and the crystal. The shielding of the photomultiplier tube is very efficient and no influence from the magnetic field in the opposite half of the instrument can be observed.

The electron-gamma arrangement is used mainly for high energy gamma-rays that are little converted and therefore less suitable for electron-electron coincidence experiments.

Limitations of the method

a. A few general considerations

Let $g(\xi/p)$ be the probability of recording an electron of momentum ξ, when the instrument is set to focus electrons of momentum p. The number of electrons recorded per unit time then becomes

$$N = N(\xi)\omega g(\xi/p)\varepsilon(\xi), \tag{1}$$

where $N(\xi)$ is the total number of electrons of momentum ξ emitted from the source per unit time. ω is the effective solid angle (effective transmission) of the instrument

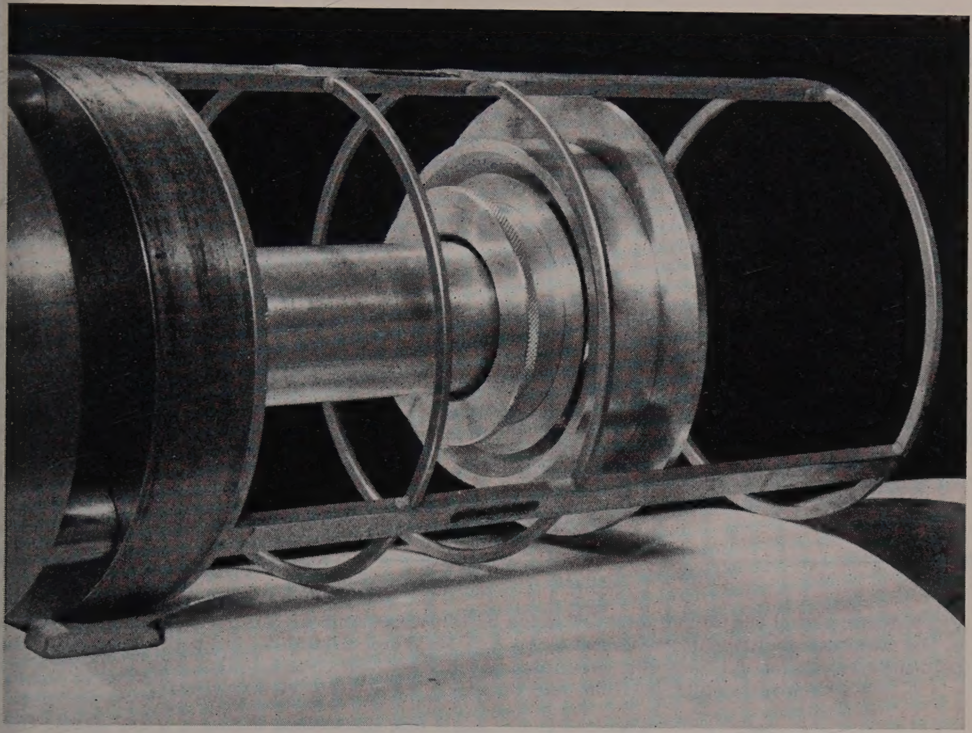

Fig. 3. Photograph showing the baffle-system in one half of the electron-electron coincidence spectrometer.

and $\varepsilon(\xi)$ is the efficiency of the detector for electrons of momentum ξ. The actual value of ω will depend on the width of the source as well as on the setting of the exit baffle B_2. Experimentally ω can easily be measured against the maximum effective solid angle Ω. To do this the peak counting rate N_{peak} of a conversion line is determined for different settings of the exit baffle. As shown in Fig. 2 the counting rate approaches a maximum $N_{\text{peak}}^{\max}$ which obviously corresponds to Ω. The effective transmission at a certain point then simply becomes

$$\omega = \Omega \frac{N_{\text{peak}}}{N_{\text{peak}}^{\max}}. \tag{2}$$

Since Ω does not depend on the width of the source it is a constant of the instrument and can be determined once and for all. This has been done in three different ways: (i) It has been geometrically computed, (ii) it has been measured with a Cs^{137} source of known absolute activity, and (iii) it has been determined by means of a coincidence method making use of the known conversion coefficient of the 569 keV transition in Pb^{207} (see below). The values agree within their limits of error and for the spectrometer considered here Ω was found to be 4.0 ± 0.3 per cent in each half.

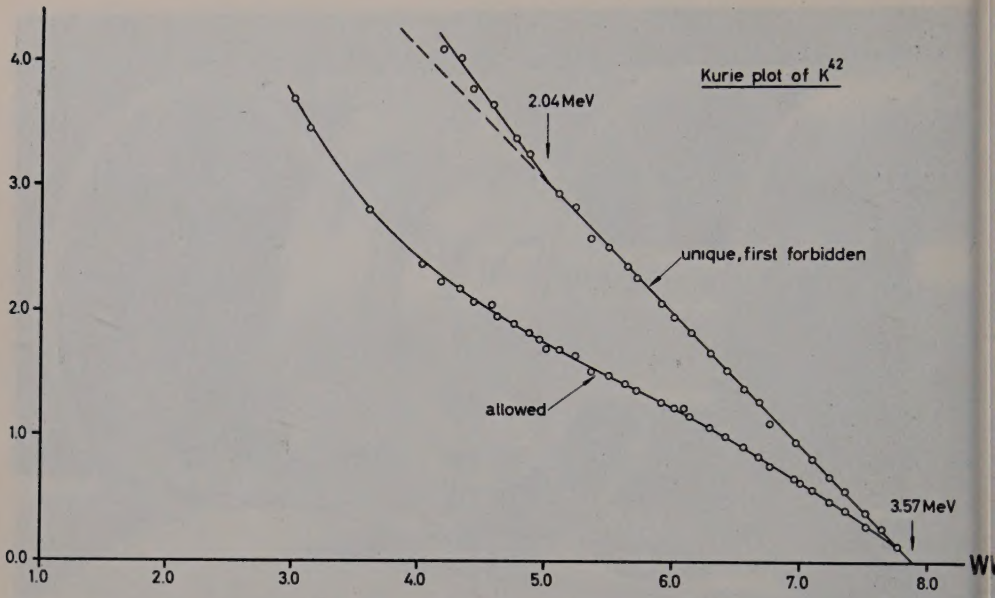

Fig. 4. Kurie plot of the unique, first forbidden beta-ray spectrum of K^{42}. The upper energy limits of the two beta components are found to be 3.57 MeV and 2.04 MeV, in good agreement with published values (cf. K. Siegbahn. Ark. Mat. Astr. Fys. *34 B*, No. 4 (1946) and L. Koertz, A. Schwartzschild, R. Gold, and C. S. Wu, Phys. Rev. *95*, 612 (1954)).

The detector efficiency $\varepsilon(\xi)$ can be determined experimentally for various electron energies by studying the shape of the continuous beta-ray spectrum. It is a wellknown fact that for an allowed spectrum corresponding to a single group of beta-rays the detector efficiency can be derived from the deviations from a straight line of the Kurie plot. For electron energies above 100 keV the efficiency is nearly 100 per cent, but below 100 keV the efficiency drops rapidly being almost zero at 40 keV. The use of scintillation detectors therefore makes it impossible to study low energy electrons.

For convenience, we introduce here the efficiency-corrected counting rates

$$ {}_{c}N(\xi) = \frac{N(\xi)}{\varepsilon(\xi)} \,. \tag{3} $$

Whenever $\xi \geq 100$ keV, ${}_{c}N(\xi) \approx N(\xi)$. Because $g(\xi/p) \neq 0$ only in the vicinity of $\xi = p$ and because $\varepsilon(\xi)$ varies slowly with ξ one can replace $\varepsilon(\xi)$ by $\varepsilon(p)$ in formula (1).

It follows from the general theory of beta-ray spectrometers that $g(\xi/p)$ depends only on the ratio ξ/p but not on p itself.[1]

Thus one has

$$ \int_0^\infty g\left(\frac{\xi}{p}\right) d\xi = \eta p \tag{4} $$

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[1] This statement holds only as long as the shape of the magnetic field is conform and independent of the absolute value of the magnetic field strength.

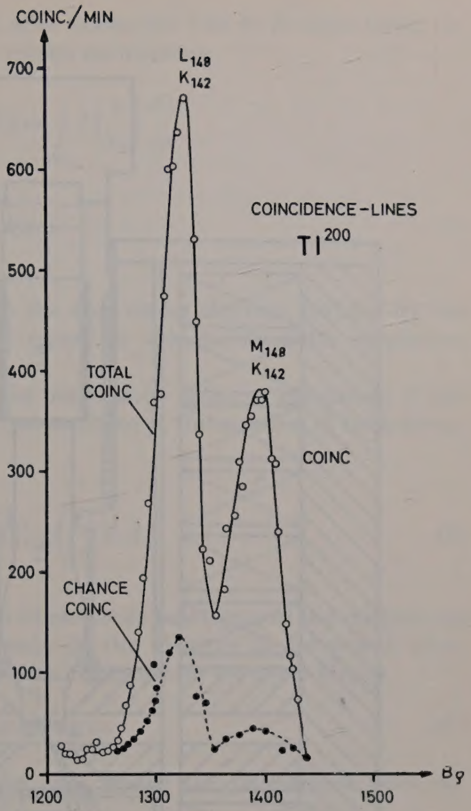

Fig. 5. Coincidence lines in Tl^{200}. One spectrometer is focusing the 142.2 keV K peak (K_{142}) while the opposite spectrometer is scanning over the strong 148.0 L and M peaks (L_{148} and M_{148}). Chance coincidences are recorded with a sufficiently large time delay introduced in one channel.

where η is a constant. $g(\xi/p)$ can easily be determined experimentally at least to an approximation since

$$g\left(\frac{\xi}{p}\right) \approx f\left(\frac{p}{p_0}\right) \tag{5}$$

where $f(p/p_0)$ is the instrumental window curve i.e. the response of the spectrometer to mono-energetic electrons. $f(p/p_0)$ is simply the shape of a conversion line obtained with a thin source. It is assumed that $f(p/p_0)$ is normalized to unity for $p = p_0$.

Let N_0 be the number of disintegrations per unit time and let δ be the probability that a certain nuclear transition takes place during the decay.[1] Finally let $\varkappa$ stand for the corresponding conversion probability.[2] If the instrument is adjusted to focus these conversion electrons the peak counting rate becomes.

$${}_cN_{\text{peak}} = N_0\,\delta\,\varkappa\,\omega. \tag{6}$$

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[1] i.e. the total feeding to this level. This is the sum of the direct beta-decay feeding and of indirect feeding caused by beta-decay is higher excited levels giving rise to the transition under consideration in the cascad down to the ground state.

[2] The conversion probability is related to the conversion coefficient: $\varkappa = \alpha(1+\alpha)^{-1}$.

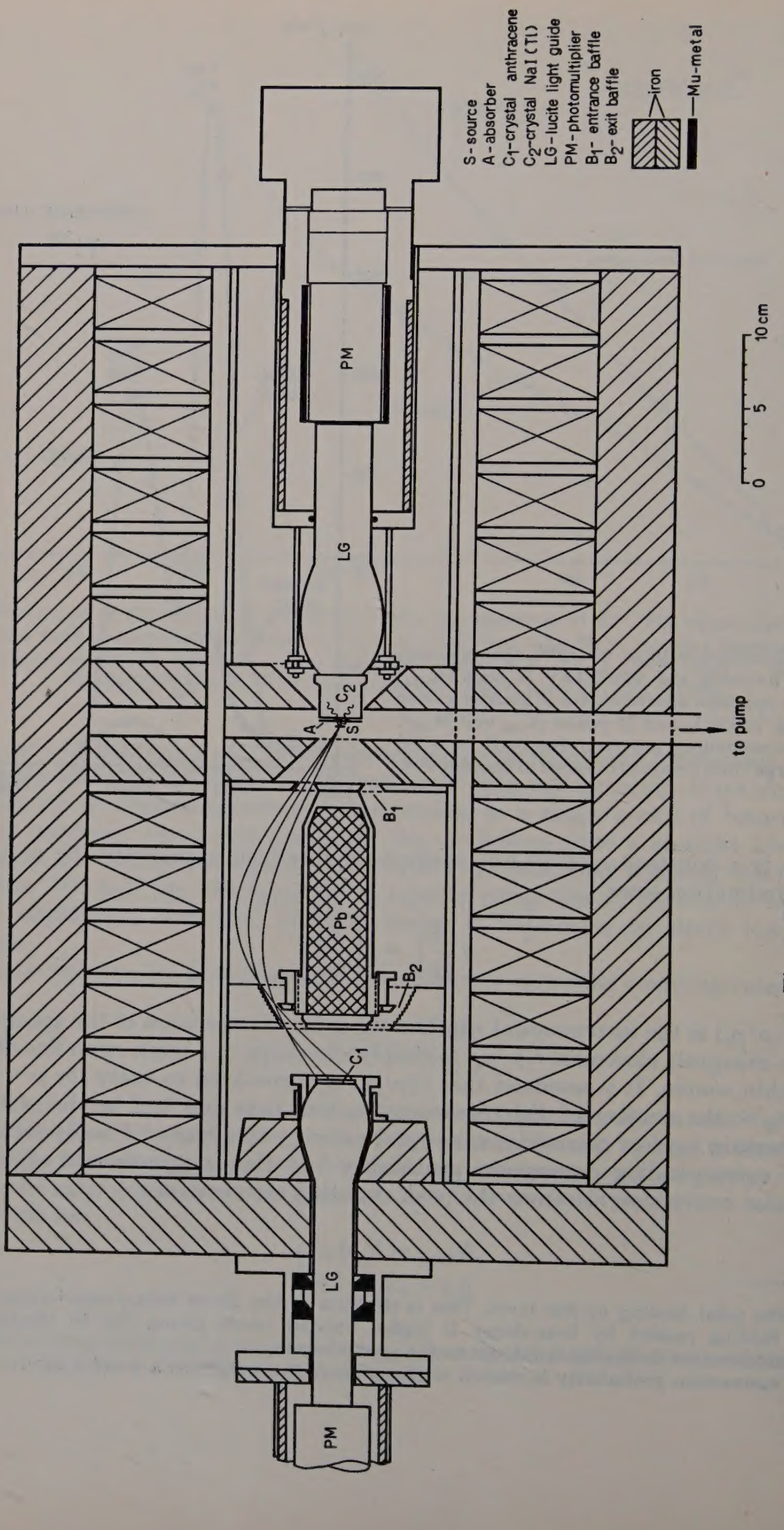

Fig. 6. Electron-gamma coincidence spectrometer.

From this expression one obtains for the area under the line in divided form, i.e. counting rate per unit momentum interval versus momentum

$$\int\limits_{\text{line}} \frac{{}_cN_{\text{line}}}{p}\,dp = N_0\,\delta\,\varkappa\,\omega \int\limits_{\text{line}} f\left(\frac{p}{p_0}\right)\frac{dp}{p}$$

or

$$\int\limits_{\text{line}} \frac{{}_cN_{\text{line}}}{p}\,dp = \eta\,N_0\,\delta\,\varkappa\,\omega. \tag{7}$$

According to (7) the constant η is equal to the area under the line, divided by the peak counting rate. This is approximately equal to the spectrometer resolution (relative halfwidth).

We shall finally consider the area under the continuous beta-ray spectrum. If the instrument is adjusted to focus electrons of momentum p, the number of beta-decay electrons recorded per unit time becomes

$${}_cN_{\text{cont}} = \int\limits_0^\infty N_0\,\varphi(\xi)\,g\left(\frac{\xi}{p}\right)\omega\,d\xi \tag{8}$$

where the function $\varphi(\xi)$ gives the distribution of electrons belonging to the continuous beta-ray spectrum. Now, since $g(\xi/p) \neq 0$ only in the vicinity of $\xi = p$ and since $\varphi(\xi)$ varies comparatively slowly with ξ, one can replace $\varphi(\xi)$ by $\varphi(p)$. Hence

$${}_cN_{\text{cont}} = N_0\varphi(p)\eta\,p\,\omega. \tag{8'}$$

From this expression one obtains for the area under the continuous beta-ray spectrum

$$\int\limits_{\text{cont}} \frac{{}_cN_{\text{cont}}}{p}\,dp = \eta\,N_0\,\omega. \tag{9}$$

b. Signal-to-noise ratio

For the sake of simplicity we shall assume, at first, that there is no interference between the conversion lines. We shall also assume that the background due to scattered electrons, gamma-rays and photomultiplier dark-current is small compared to the corresponding channel counting rates. A discussion will be given below of what happens, when these conditions are not fulfilled. Further, we make the simplifying assumption, that the time delay between the members of a genuine coincidence pair is short compared to the resolving time of the coincidence circuit. The situation that occurs, when the delay is comparable to, or longer than, the coincidence resolving time will be discussed below.

Apart from these restrictions we shall consider a complex nuclear disintegration quite generally, and not introduce any specific assumptions concerning the decay scheme.

Suppose that spectrometer 1 is adjusted to select conversion electrons corresponding to the nuclear transition 1 and that spectrometer 2 is focusing conversion electrons corresponding to the transition 2. Let ${}_cN_1$ and ${}_cN_2$ be the counting rates in channel

1 and channel 2 respectively and let ${}_cN_g$ and ${}_cN_{ch}$ be the genuine and random coincidence counting rates respectively. All counting rates are efficiency corrected according to (3). ε_c is the efficiency of the coincidence equipment which is generally adjusted to 100 per cent. γ, finally, is a coincidence correlation constant. This takes into account, that the probability of detecting a certain electron in one spectrometer can be slightly influenced by the fact that it is in coincidence with an other electron recorded in the opposite spectrometer. This can be due to angular correlation effects, to misalignments of the source or to absorbtion of the electrons in the source itself. For properly centered sources of good specific activity γ is equal to unity, since the angular correlation, if present, will tend to spread over the 30° aperture angle in each half of the instrument (cf. Fig. 1).

One then obtains the following general expressions

$$ {}_cN_1 = N_0\varphi(p_1)\eta_1 p_1\omega_1 + N_0\delta_1\varkappa_1\omega_1, \tag{10}$$

$$ {}_cN_2 = N_0\varphi(p_2)\eta_2 p_2\omega_2 + N_0\delta_2\varkappa_2\omega_2, \tag{11}$$

$$ {}_cN_g = \{N_0\delta_1\varkappa_1\omega_1\delta_2\,{}_1q_2\varkappa_2\omega_2 + N_0\varphi(p_1)\eta_1 p_1\omega_1\delta_2\,{}_\beta q_2\varkappa_2\omega_2 + N_0\varphi(p_2)\eta_2 p_2\omega_2\delta_1\,{}_\beta q_1\varkappa_1\omega_1\}\varepsilon_c\gamma, \tag{12}$$

$$ {}_cN_{ch} = 2\,\tau\,{}_cN_1\,{}_cN_2. \tag{13}$$

2τ is the resolving time of the coincidence circuit.

In formula (12) we have introduced the coincidence coupling constants ${}_1q_2$, ${}_\beta q_1$ and ${}_\beta q_2$. These are defined as follows. Let ${}_1w_2$ be the probability per nuclear disintegration, that the transition 1 will follow (or be followed by) the transition 2. Then

$$ {}_1w_2 = \delta_1\delta_2\,{}_1q_2. \tag{14}$$

By analogy ${}_\beta w_1$ and ${}_\beta w_2$ are the probabilities per nuclear disintegration that transition 1 and transition 2, respectively, will follow an electron belonging to the fraction of the continuous beta-ray spectrum accepted by the opposite spectrometer. Thus

$$ {}_\beta w_1 = \varphi(p_1)\eta_1 p_1\delta_2\,{}_\beta q_2, \tag{15}$$

$$ {}_\beta w_2 = \varphi(p_2)\eta_2 p_2\delta_1\,{}_\beta q_1. $$

It is convenient to introduce the following relations

$$ \chi_1 = \frac{n_{1\beta}}{n_{1e}} = \frac{\varphi(p_1)\,p_1\,\eta_1}{\delta_1\,\varkappa_1} \tag{16}$$

and

$$ \chi_2 = \frac{n_{2\beta}}{n_{2e}} = \frac{\varphi(p_2)\,p_2\,\eta_2}{\delta_2\,\varkappa_2} \tag{17}$$

where n_β is the number of electrons belonging to the continuous beta ray spectrum and n_e is the number of conversion electrons recorded per unit time in the corresponding channels. χ_1 and $\chi_2 \lesssim \eta_1/\delta_1\varkappa_1$ and $\eta_2/\delta_2\varkappa_2$ respectively, since it can be shown [9] that $\varphi(p)p \lesssim 1$.

From (12) and (13) one gets for the ratio of genuine to random coincidences—i.e. the signal to noise ratio

$$\frac{{}_cN_g}{{}_cN_{ch}} = \frac{\varepsilon_c \gamma}{2\,N_0 \tau} \, \frac{{}_1q_2 + {}_\beta q_2 \chi_1 + {}_\beta q_1 \chi_2}{1 + \chi_1 + \chi_2 + \chi_1 \chi_2} \tag{18}$$

or

$$\frac{{}_cN_g}{{}_cN_{ch}} = \frac{\varepsilon_c \gamma}{2\,N_0 \tau} R \tag{18'}$$

where R is the *coincidence coupling ratio.* This ratio can be larger as well as smaller than unity depending on the coincidence coupling constants.

Considerations of this kind have been published, but so far the treatments have been limited to simple decay schemes. For instance one has considered the case of electron capture followed by two gamma-rays in cascade. In that case one has $R = 1$. However, the necessity of taking into account the coincidence coupling ratio when considering the possibilities of the electron-electron coincidence spectrometer must be stressed. It is easily seen, that under less favourable conditions the signal to noise ratio can be reduced by orders of magnitude.

The signal to noise ratio will also be affected by the presence of a *background* caused by scattered electrons, gamma-rays or photomultiplier dark-current. Interference with neighbouring conversion lines will give rise to the same effect.

To take the influence of the background into account in the expression for R one should replace χ_1 and χ_2 in the *denominator*[1] of (18) with χ_1' and χ_2' defined in analogy with (16) and (17) as

$$\chi_2' = \frac{n_{1\beta} + n_{1\,\mathrm{backgr}}}{n_{1e}} \tag{16'}$$

and

$$\chi_2' = \frac{n_{2\beta} + n_{2\,\mathrm{backgr}}}{n_{2e}}. \tag{17'}$$

The coincidence coupling ratio then becomes

$$R = \frac{{}_1q_2 + {}_\beta q_2 \chi_1 + {}_\beta q_1 \chi_2}{1 + \chi_1' + \chi_2' + \chi_1' \chi_2'}. \tag{19}$$

Studies of nuclear decay schemes

The first task that presents itself in all studies of nuclear disintegrations is the determination of the decay scheme. In simpler cases this can be done by ordinary scintillation or beta-ray spectrometer methods. For more complex nuclear disintegrations, however, it becomes necessary to resort to coincidence technique in order to get a unique decay scheme. It is evident, that the good energy resolution in both channels provided by the electron-electron coincidence spectrometer makes it very suitable for this kind of experiments.

a. e^--e^- coincidences

For conversion-electrons following electron capture such experiments are readily performed. The two spectrometers are adjusted to focus the peaks of the conversion lines. One then has

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[1] We assume, that the background signals do not give rise to any genuine coincidences.

$$R_{12} = {}_1q_2 \tag{19'}$$

and

$$_cN_g = N_0 \delta_1 \varkappa_1 \omega_1 \delta_2 \, {}_1q_2 \varkappa_2 \omega_2. \tag{12'}$$

$_cN_g > 0$ means $q > 0$ and proves that the transitions under investigation belong to the same branch of the decay. Because of the good resolution the coincidence assignment generally becomes unique. Sometimes, however, there may be an interference between different lines. Nevertheless, from a comparison between the shape of the coincidence spectrum and the gross spectrum in the corresponding channel one can generally decide to which transitions the coincidence coupling should be referred.

When the conversion lines are superimposed on an intense background or on the continuous beta-ray spectrum the importance of a good resolution is even more pronounced. That is so because the coincidence coupling ratio depends on the spectrometer resolution as seen from formulae (16′) and (17′).

It is often convenient to use the off-set method in order to obtain from the total number of genuine coincidences, the contribution due to e^--e^- coincidences only. For this purpose three different quantities are measured.

(1) The number of genuine coincidences per unit time obtained with both spectrometers set at the peaks of the two lines 1 and 2: $N_g(\text{tot})$.

(2) The number of genuine coincidences per unit time obtained with spectrometer 1 set just above the high energy edge of line 1 and spectrometer 2 at line 2: $N_g(\beta e^-)$.

(3) The number of genuine coincidences per unit time obtained with spectrometer 1 at line 1 and spectrometer 2 set just above the high energy edge of line 2: $N_g(e^-\beta)$.

One then has

$$_cN_g(e^-e^-) \approx {}_cN_g(\text{tot}) - \{ {}_cN_g(\beta e^-) + {}_cN_g(e^-\beta)\}. \tag{20}$$

b. *β-e⁻-coincidences*

It is easily seen that for coincidence measurements between beta-rays and conversion electrons a low resolution–high transmission setting should be used in the beta-channel in order to increase the coincidence counting rate. In the opposite channel, however, good resolution should be maintained, since

$$R_{\beta e} = \frac{{}_\beta q_1}{1 + \chi_1'} \tag{19''}$$

and

$$_cN_g = N_0 \varphi(p_1) \eta_1 p_1 \omega_1 \delta_2 \, {}_\beta q_2 \varkappa_2 \omega_2. \tag{12''}$$

For $\chi_1' = 1$ the coincidence coupling ratio is not too sensitive to the resolution, but under less favourable conditions the signal to noise ratio will be considerably reduced with reduced resolving power. It can therefore be advisable to increase the resolution, at the expense of the transmission, since R varies as η^{-1} but ω only as $\eta^{\frac{1}{2}}$ according to the theory of long lens spectrometers with ring focus baffles.

c. *Applications*

$_cN_g > 0$ implies ${}_1q_2 > 0$, where q is the corresponding coincidence coupling constant. From coincidence information of this kind it is often possible to determine the decay scheme uniquely. In particular, this is true if the coincidence data are combined with precise determinations of the transition energies obtained from measurements of the conversion lines with a high resolution instrument. From the latter experiments, one obtains sum-up combinations, e.g. cascade transitions with cross-over gamma-rays corresponding to the same total transition energy. These sum-up combinations provide additional information concerning the branching of the decay.

The coincidence spectrometer has been used to study the decay schemes of the following neutron deficient isotopes.[1]

$$\mathrm{Bi}^{205} \xrightarrow[(14.5\ \mathrm{d})]{} \mathrm{Pb}^{205} \qquad \text{(ref. [10])}$$

$$\mathrm{Pb}^{200} \xrightarrow[(21\ \mathrm{h})]{} \mathrm{Tl}^{200} \qquad \text{(ref. [11])}$$

$$\mathrm{Tl}^{200} \xrightarrow[(27\ \mathrm{h})]{} \mathrm{Hg}^{200} \qquad \text{(ref. [12])}$$

$$\begin{tikzcd}
\mathrm{Pb}^{202\,\mathrm{m}} \arrow[r, "(3.7\ \mathrm{h})"'] \arrow[d] & \mathrm{Tl}^{202\,\mathrm{m}} \arrow[d] \\
\mathrm{Pb}^{202} \arrow[r] & \mathrm{Tl}^{202}
\end{tikzcd} \qquad \text{(ref. [13])}$$

$$\mathrm{Hg}^{195\,\mathrm{m}} \xrightarrow[(40\ \mathrm{h})]{} \mathrm{Au}^{195} \qquad \text{(ref. [14])}$$

$$\begin{tikzcd}
\mathrm{Pb}^{197\,\mathrm{m}} \arrow[r, "(42\ \mathrm{m})"'] \arrow[d] & \mathrm{Tl}^{197} \\
\mathrm{Pb}^{197} &
\end{tikzcd} \qquad \text{(ref. [15])}$$

Details of the decay schemes are discussed in the references. The coincidence coupling information obtained for the decay schemes shown in Figs. 7, 8 and 9 are given in Tables 1, 2 and 3 respectively.

Measurements of internal conversion coefficients

The internal conversion coefficients are of considerable interest, since they depend on the multipolarity and character of the nuclear transition. They can therefore be used in the determination of spins and parities of nuclear levels. Theoretical calculations [16] are now available, that take into account screening and finite nuclear size effects. They are supposed to be correct to within a few per cent. This implies that precise measurements of K-conversion coefficients can be used also to determine the mixing ratios in transitions of mixed electric and magnetic character. The internal conversion coefficient, the multipolarity and character as well as the mixing ratio must be known in order to make possible a deduction of the partial electromagnetic transition probabilities from the experimental halflife of an excited level (cf. next section).

a. Conventional methods

In principle the internal conversion coefficients can be measured in different ways with the aid of a scintillation spectrometer. Under favourable conditions and with great care accuracies of 5–10 per cent have been achieved [17]. Generally, however,

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[1] The halflives of the mother-activities are given inside the brackets.

Table 1. Coincidence coupling information on Tl^{200}.

Electron-electron coincidence measurements

Coinc. obs.	No. coinc. obs.
148.0–109.5	148.0–158.9
148.0–142.2	148.0–257.3
148.0–235.3	148.0–289.5
148.0–268.0	

Table 2. Coincidence coupling information on Hg^{200}.

Coinc. obs.	No coinc. obs.
Electron-electron coincidence measurements	
252.0– 289.0 (?)	252.0– 309.2
252.0– 368.0	252.0– 579.4
289.0– 368.0	252.0– 785.3
289.0–1226.9	252.0– 828.6
309.2– 368.0	252.0–1206.7 + 1226.9
309.2–1206.7	252.0–1362.3 (?)
368.0– 579.4	352.0–1516.7 (?)
368.0– 828.6	289.0–1206.7
368.0–1206.7 + 1226.9	309.2– 828.6
368.0–1362.3	309.2–1226.9
368.0–1516.7	785.3– 828.6
579.4– 712.1 (?)	
579.4– 785.3	
579.4– 828.6	
Electron-gamma coincidence measurements	
Coinc. obs.	
252.0–1000 (γ)	
252.0– 368.0 (γ)	
579.4– 368.0 (γ)	

the uncertainties in the internal conversion coefficients obtained with scintillation spectrometers are considerable. The decay scheme must be completely known and should not be too complex.

More accurate measurements of internal conversion coefficients are possible with a beta-ray spectrometer. Two different methods have been developed. One possibility [18, 19, 20] is to determine the ratio between the number of conversion electrons recorded per unit time and the number of secondary electrons recorded per unit time expelled from a suitable converter. The efficiency of the converter for the gamma-ray energy of interest should be accurately known. This efficiency depends not only on the absolute photo-electric cross-section at the energy in question, but also on the angular distribution of the photoelectrons as well as on straggling effects in the

Table 3. Coincidence coupling information on Pb^{205}.

Coinc. obs.	No. coinc. obs.	
Electron-electron coincidence measurements		
260.5–282.3 + 282.2	260.5–987.8	284.2–1766.4
260.5–349.4	260.5–1043.7	349.4–580.0
260.5–571.0	260.5–1766.4	349.4–987.8
260.5–703.3 (?)	260.5–1777.4	349.4–1043.7
282.3–284.2 (?)	282.3–349.4	511.7–703.3
282.3 + 284.2–511.7	282.3–1043.7	571.0–703.3
282.3 + 284.2–571.0	284.2–349.4	703.3–987.8
284.2–703.3	284.2–910.8	703.3–1043.7
349.4–571.0	284.2–1043.7	
349.4–703.3		
571.0–580.0 (?)		
703.3–910.8		
Electron-gamma coincidence measurements		
260.5–1614.6 (γ)	260.5–1766.4 (γ)	571.0–1766.4 (γ)
284.2–703.3 (γ)	260.5–1777.4 (γ)	571.0–1777.4 (γ)
703.3–1863.3 + 1906.5 (γ)	260.5–1863.3 (γ)	703.3–1766.4 (γ)
703.3–annih rad (γ)	260.5–1906.5 (γ)	703.3–1777.4 (γ)
(?)	282.3–1766.4 (γ)	987.8–1614.6 (γ)
	282.3–1774.4 (γ)	987.8–1766.4 (γ)
	282.3–1863.3 (γ)	987.8–1777.4 (γ)
	282.3–1906.5 (γ)	987.8–1868.3 (γ)
	284.2–1766.4 (γ)	987.8–1906.5 (γ)
	284.2–1777.4 (γ)	1043.7–1766.4 (γ)
	284.2–1863.3 (γ)	1043.7–1777.4 (γ)
	284.2–1906.5 (γ)	1043.7–1863.3 (γ)
	349.4–1766.4 (γ)	1043.7–1906.5 (γ)
	511.7–1766.4 (γ)	

converter material. Therefore it is difficult to calculate the efficiency of the converter theoretically. Alternatively these efficiencies can also be determined experimentally.

The other method consists of a comparison between the area under the conversion line and the area under the continuous beta-ray spectrum. This is considered to be the most accurate method, at present, for determination of internal conversion coefficients. According to (7) and (9) one has

$$a_0 = \frac{\int\limits_{\text{line}} \frac{{}_{c}N_{\text{line}}}{p}\,dp}{\int\limits_{\text{cont}} \frac{{}_{c}N_{\text{cont}}}{p}\,dp} = \delta_{\text{line}}\,\varkappa. \tag{21}$$

Thus

$$\varkappa = \frac{1}{\delta_{\text{line}}}\,a_0. \tag{21'}$$

The method is limited to the most simple beta-decays, where δ_{line} can be obtained from an analysis of the various components of the beta-ray spectrum, reproduced in a Kurie plot.

The conversion coefficient is related to the conversion probability according to the formula

$$\alpha = \frac{\varkappa}{1-\varkappa}. \tag{22}$$

From (22) one has

$$\frac{\Delta\alpha}{\alpha} = \frac{1}{1-\varkappa}\,\frac{\Delta\varkappa}{\varkappa}. \tag{23}$$

Evidently, α will be determined inaccurately in this way, when $\varkappa \approx 1$. In most practical cases, however, $\varkappa$ is of the order of per cent, or smaller.

b. The coincidence spectrometer method[1]

Suppose spectrometer 1 is focusing line 1, while spectrometer 2 is scanning line 2. For the sake of simplicity we will assume that $\eta_1 = \eta_2 = \eta$. From (7) and (12) one obtains

$$a_1 = \frac{\displaystyle\int\limits_{\substack{\text{line 2}\\ \text{coinc 1}}} \frac{{}_cN_g}{p_2}\,dp_2}{\displaystyle\int\limits_{\text{line 2}} \frac{{}_cN_2}{p_2}\,dp_2} = \delta_{1\,2}\, q_1\, \varkappa_1\, \omega_1\, \varepsilon_c\, \gamma. \tag{24}$$

This expression can be used at once to determine the internal conversion probability provided the spectrometer has been calibrated in regard to transmission as discussed above and that $\varepsilon_c\gamma$ is known. Generally $\varepsilon_c\gamma$ can be put equal to unity if the source is properly centered and of good specific activity, and if the coincidences can be considered prompt.

Obviously expression (24) can also be used in determining the transmission—or the coincidence coupling probability—if the conversion coefficient is known. Alternatively one can use the continuous beta-ray spectrum to calibrate the instrument directly. According to (9) and (12), if spectrometer 2 is kept fixed on line 2, when spectrometer 1 is scanning the spectrum, one has

$$a_2 = \frac{\displaystyle\int\limits_{\substack{\text{cont 1}\\ \text{coinc 2}}} \frac{{}_cN_g}{p_1}\,dp_1}{\displaystyle\int\limits_{\text{line 2}} \frac{{}_cN_2}{p_2}\,dp_2} = \omega_1\, \varepsilon_c\, \gamma. \tag{25}$$

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[1] A method similar to that described in this section has been developed at Zürich [21]. The Swiss group uses a single long lens spectrometer operated in coincidence with a scintillation spectrometer.

We assume γ to be constant.

The coincidence coupling constant vanishes in the integral, because all transitions 2 will follow a beta decay. From (24) and (25) we finally get

$$\varkappa_1 = \frac{1}{\delta_{1\,2}q_1}\frac{a_1}{a_2}. \tag{26}$$

If (26) is compared to (21′) it is seen that the coincidence spectrometer method is the more general. δ_{line} is the total feeding to this transition. This includes not only the direct beta-decay feeding, but also feedings to higher excited states, giving rise to this transition in the cascade down to the ground state. $\delta_{1\,2}q_1$, however, is, according to (14), the probability per transition 2 for that transition to take place *in coincidence with 1.* This probability is known in many cases, where the complete decay scheme is unknown. For instance $\delta_{1\,2}q_1 = 1$ whenever transition 1 is a transition from the first excited state to the ground state, independently of the actual structure of the decay scheme as a whole.

The coincidence method can further be applied as well to cases of electron capture and positron decay as to the study of long-lived isomers, because different sources, or rather, different isotopes present in the same source, can be used to measure the a_1 and a_2 ratios. As is easily seen branching ratios can also be measured by means of the coincidence spectrometer method.

The most serious limitation encountered in this technique is due to the fact that the coincidence counting rate sometimes becomes inconveniently low. Therefore the accuracy of the values obtained is generally limited by the statistical uncertainties.

c. *Applications*

The coincidence method has been employed to get information about internal conversion coefficients and branching ratios for the decay schemes shown in Figs. 7, 8 and 9. A few examples are given here to illustrate the procedure.

A 2 × 3 mm source of Bi^{207} deposited on a 2–3 mg/cm^2 copper backing was introduced into the spectrometer. The exit baffles were opened 5 turns. The counting rate at the peak of the 1064 keV K-line was 19800 cpm. The 569 keV line was focused in the opposite spectrometer. The genuine coincidence counting rate was then found to be 8 ± 1 coinc./min. In this case one has $\delta_{1\,2}q_1 = 1$ since it is known, that the 569 keV transition leads from the first excited state to the ground state. The K-conversion coefficient for this transition has been measured [22] to be 1.7 per cent, which is in accordance with the theoretical value [16] for an electric quadrupole transition.

For these high energy lines $\varepsilon_c\gamma = 1$. This means that the results obtained with the coincidence instrument can be used for determining the effective transmission of the spectrometer focusing the 569 keV line. According to (24) one has

$$\omega_1 = \frac{8 \pm 1}{19\,800 \cdot 1.7 \times 10^{-2}} = (2.4 \pm 0.3) \times 10^{-2}.$$

We use here the peak counting rates instead of the areas under the line profiles.

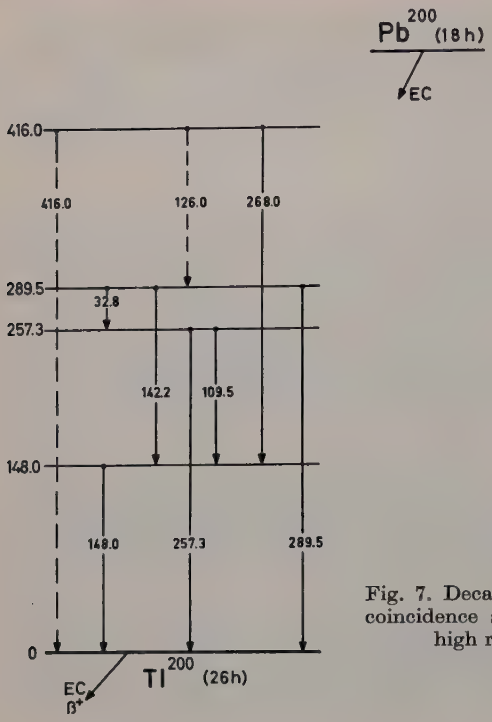

Fig. 7. Decay scheme of Pb^{200}, based on information from coincidence spectrometer measurements (cf. Table 1) and high resolution determinations. See refr. [11].

This value is in agreement with that found from Fig. 2 for the effective transmission in this case. The measurement therefore serves as a check of the maximum transmission calibration.

The diameter of the source used in the study of Tl^{200} was 2 mm. The exit baffles were opened 6 turns corresponding to a transmission of about 3.3 per cent (cf. Fig. 2). The decay scheme, shown in Fig. 8, is based on coincidence coupling information and high resolution measurements. According to this scheme transitions from higher excited states to the ground state take place only via the 368 keV transition from the first excited state to the ground state. One would then expect to get the same a_1 ratios for all conversion lines, when they are measured in coincidence with the 368 keV *K*-line.

The 368 keV *K*-line was tried in coincidence with eight different lines belonging to this decay. It was found to be in coincidence with all of them (cf. Table 2). For seven[1] of these coincidence combinations the a_1 ratios were found to be the same within the limits of the statistical errors. This proves that the decay to the ground state takes place mainly via the first excited state. From the measurements a_1 was found to be $(11 \pm 1) \times 10^{-4}$. According to (24) this corresponds to a conversion prob-

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[1] The coincidence counting rate obtained with the 252 keV *K*-line was somewhat too low. If this result is significant, there should be a cross-over transition from the 1884.7 keV level directly to the ground state. However no conversion lines have been observed corresponding to such a transition. The coincidences obtained with the 252- and 368-lines were found to be prompt compared to the moderate resolving time of the coincidence circuit used in these experiments ($2\,\tau_0 = 0.15$ µsec.).

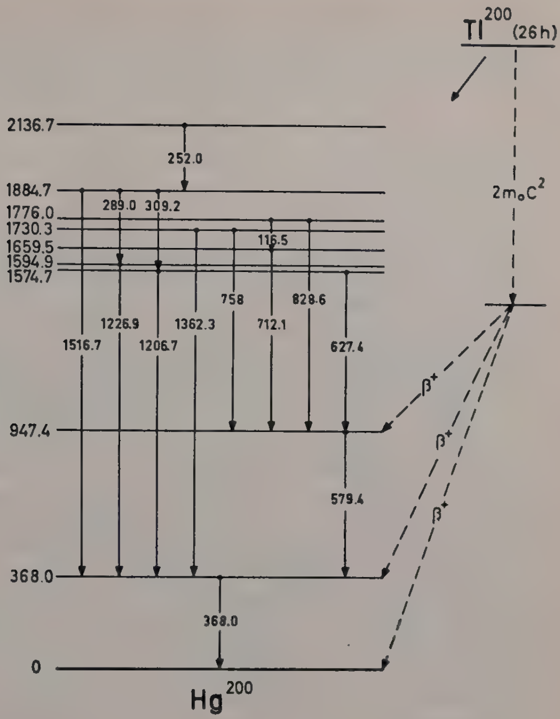

Fig. 8. Decay scheme of Tl^{200}, based on information from coincidence spectrometer measurements (cf. Table 2) and high resolution determinations. See refr. [12]. (*Erratum:* 758 should be 785.3.)

ability of (3.3 ± 0.3) per cent or a conversion coefficient of (3.4 ± 0.3) per cent. The theoretical value [16] for a pure electric quadrupole transition of 368 keV is $\alpha_2 = 3.57 \times 10^{-2}$.

The coincidence measurements on Bi^{205} (cf. Fig. 9) were made with a transmission of 2.5 per cent in each channel. As shown in Table 3 the 703.3 transition was found to be in coincidence with the 260.5– (?), 349.4–, 284.2 and 910.8 keV transitions. The 260.5 keV transition is not accounted for in the decay scheme. The remaining three transitions, however, should, according to the decay scheme, all be in coincidence to 100 per cent, i.e. $\delta q = 1$. With this assumptions and with $\varepsilon_c \gamma = 1$, the internal conversion coefficients can be determined from the experimental data. The results are

284.2–703.3	$\alpha_{703.3} = (8.4 \pm 1) \times 10^{-3}$
349.4–703.3	$\alpha_{703.3} = (11.0 \pm 3) \times 10^{-3}$
910.8–703.3	$\alpha_{703.3} = (7.5 \pm 2.5) \times 10^{-3}$

These figures should be compared to the theoretical value [16] for a pure electric quadrupole transition of 703 keV.

$$\alpha_2 = 11 \times 10^{-3}.$$

Further applications of this method are discussed in references [10] and [12].

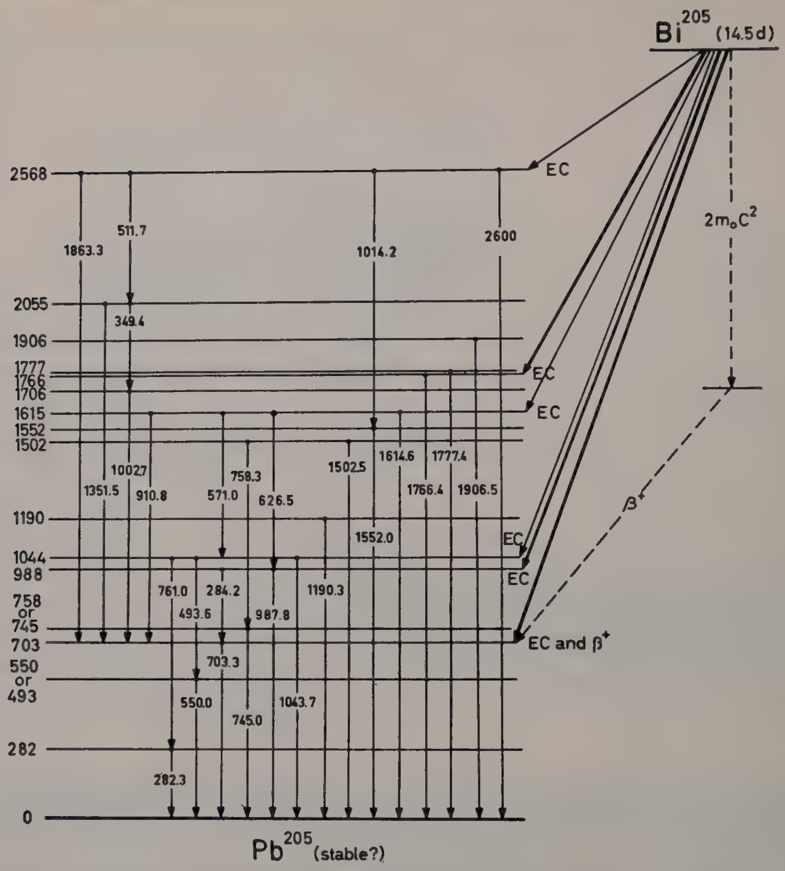

Fig. 9. Decay scheme of Bi^{205}, based on information from coincidence spectrometer measurements (cf. Table 3) and high resolution determinations. See refr. [10].

Lifetime measurements

a. Delayed coincidence method

We have so far assumed that the delay between the two members of a coincidence pair is short compared to the resolving time of the coincident circuit. This is generally the case when the coincidence equipment is operated with a moderate resolving time of about a few tenths of a microsecond. If, however, a millimicrosecond coincidence unit is used instead, the lifetimes of the excited nuclear levels will be comparable to the coincidence resolving time, at least for low energy transitions. It then becomes necessary to replace the efficiency ε_c of the coincidence circuit with the probability, that the two events take place within the coincidence resolving time.

If $y = x - t$, one has according to Newton [23] for this probability

$$F(x) = \lambda e^{-\lambda x} \int_{\infty}^{x} e^{\lambda y} P(y)\, dy. \qquad (27)$$

x stands for an artificial time delay introduced in one channel and λ is the decay constant of the excited state under consideration and $P(x)$ is a prompt coincidence curve recorded under otherwise identical conditions. From (27) one obtains

$$\frac{d}{dx}(\ln F(x)) = -\lambda\left\{1 - \frac{P(x)}{F(x)}\right\}. \tag{28}$$

$F(x)$ is the delayed coincidence curve. There is always a region where $P(x) \ll F(x)$, if the resolving time of the coincidence circuit is considerably shorter than the lifetime of the excited state. It is seen from formula (28) that under such conditions λ can easily be determined from the slope of the curve $\ln F(x)$ versus x. When the lifetime is comparable to, or shorter than, the resolving time, λ can sometimes be obtained from the shape of the whole curve by means of a centroid shift analysis, as first shown by Bay [24]. Whenever this method is applied, however, particular care should be taken to avoid systematic errors.

The sequence of excited levels and their spins and parities generally depend on rather gross parameters of the nuclear model. The electromagnetic transition probabilities, however, reveal finer details about the nuclear structure. The transition rates are proportional to the squares of the corresponding matrix elements, containing the wave functions of the initial and final states as well as the appropriate transition operator. Measurements of the halflives of excited states, therefore, provide sensitive tests for the nuclear model. The measured halflives should be corrected for branching, internal conversion and mixing. These corrections can be determined with the coincidence spectrometer method as discussed above. Thus the coincidence spectrometer can be used for measuring the halflives of specified excited levels and for determining the electro-magnetic transition rates, even in very complex nuclear disintegrations.

However the present detector arrangement is not suitable for the shortest resolving times and there are plans to reconstruct the detectors. The cylindrical parts of the lightguides will be omitted and the photomultiplier tubes will be placed closer to the crystal in order to reduce the transit time spread of the photons. RCA C-7187 A development tubes will be used (endcathod, gain at 2500 volt 5×10^7). Preliminary experiments with these tubes indicate a coincidence resolving time $2\tau \sim 10^{-9}$ sec at an efficiency of about 90 per cent and at energies of about 100 keV dissipated in the scintillators.

b. High frequency method

We will finally add a few words about a possible method for lifetime measurements, which has been suggested by Gerholm and Tove [25]. The method should be particulary suitable for fast, low energy transitions. These cannot be measured with the present delayed coincidence method. Suppose spectrometer 1 is focusing line 1 and spectrometer 2 is scanning line 2. The shape of the coincidence line will be equal to the shape of the corresponding single channel line, but scaled down by a factor a_1 (cf. formula 24).

If the source is connected to an alternating voltage of high frequency the conversion lines will wobble with the frequency of the transmitter. If spectrometer 1 is kept constant on its original setting it will accept only those electrons that have not undergone a change of momentum. It is easily seen that the coincidence line will be broadened, when λ is comparable to the frequency of the alternating voltage Since the window curve, the amplitude of the alternating voltage and the frequency are all known it would be possible to determine the lifetimes from the broadening of the coincidence line.

An estimate of the shortest lifetimes that could be measured with this method can be made as follows. The time dependence of the electron momenta can be expressed by

$$p = p_0 + \overline{p} \sin \omega t. \tag{29}$$

In the first approximation one has

$$\eta = \frac{\Delta p}{p} = \frac{\overline{p}\,\omega\,\Delta t \cos \omega t}{p_0 + \overline{p} \sin \omega t}$$

or

$$\Delta t = \frac{\eta \{ p_0 + \overline{p} \sin \omega t \}}{\omega\,\overline{p} \cos \omega t}. \tag{30}$$

At $t = 0$ this expression gives

$$\Delta t \approx \frac{\eta\,p_0}{\omega\,\overline{p}}. \tag{30'}$$

If one now assumes

$$\begin{aligned} \eta &= 2 \times 10^{-2} \\ p_0 &= 1120\,B\varrho (= 100 \text{ keV}) \\ \omega &= 3 \times 10^{9} \text{ cps} \\ \overline{p} &= 24\,B\varrho\ (\sim 4 \text{ kV}) \end{aligned}$$

one has

$$\Delta t \approx 3 \times 10^{-10} \text{ sec.}$$

This is at least an order of magnitude shorter then the shortest resolving times tha could be obtained with the delayed coincidence method. At lower energies the hig frequency method should be even more favourable. It should also be observed, tha the resolving time of the coincidence unit is of importance only for the signa to noise ratio. Therefore G.M. counters could be used provided the source strengt is reduced in proportion to the longer resolving time. This means, that very lo energy electrons could be studied. For instance it should be possible to use th auger electrons following electron capture in coincidence with conversion electrons

On the other hand, for obvious reasons, the method is limited to low energies an to cases, where high coincidence counting rates can be achieved. Experiments alon these lines are now being performed. The method will be discussed in full in a forthcoming publication.

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